



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE  
**Board of Patent Appeals and Interferences**

APPELLANTS: Chunshan Song, Jian-Ping ) PATENT APPLICATION  
Shen, Lawrence D. Lillwitz )  
APPLICATION NO.: 09/771,876 ) Group Art Unit: 1725  
FILED: January 29, 2001 ) Examiner: Christina A.  
Ildebrano )  
FOR: SELECTIVE METHYLATION ) Attorney Docket No.:  
CATALYST, METHOD OF 38058  
CATALYST MANUFACTURE AND  
METHYLATION PROCESS )

**APPEAL BRIEF**

Commissioner  
for Patents  
Washington, D.C. 20231

<u>CERTIFICATE OF MAILING</u>	
I HEREBY CERTIFY THAT THIS CORRESPONDENCE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE WITH SUFFICIENT POSTAGE AS FIRST CLASS MAIL IN AN ENVELOPE ADDRESSED TO: COMMISSIONER FOR PATENTS, WASHINGTON, D.C. 20231 ALEXANDRIA, VA	
on 6-9	2004 By Carol A. Wilson
	(Signature)
Carol A. Wilson	
(Typed or Printed Name of Person Signing Certificate)	

This document is an appeal from the Final Rejection dated July 9, 2003, regarding the above-mentioned case. A copy of the appealed claims is attached to this brief in Appendix A.

**Real Party in Interest**

The subject patent application has been assigned to BP Corporation North America Inc.

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**ANY ADDITIONAL FEES REQUIRED  
CHARGE TO DEPOSIT ACCOUNT  
NO. 01-0528**

### Related Appeals and Interferences

Appellants are not aware of any other appeals or interferences which will directly affect or be affected or have a bearing on the Board's decision in this pending appeal.

### Status of Claims

Claims 1-8, 10, 11, 13-19, 21-23, 25, 26, 39, 40, 42, and 43 remain pending and have been finally rejected by the Examiner. In the amendment submitted with this Appeal Brief, Claims 15, 40, and 44 have been cancelled in rewritten as independent claims 47, 48, and 49 to be deemed allowable per the Examiner's instruction. Claims 9, 12, 20, 24 and 27 have been cancelled. Claims 45 and 46 have been allowed.

### Status of Amendments

An amendment has been filed along with this Appeal Brief as a separate paper. The amendment has not been acted on by the Examiner. Otherwise, there are no other pending or outstanding amendments.

### Summary of the Invention

The subject matter set forth in this patent application and regarded by Appellants as the invention is a catalyst for methylating a naphthalenic feedstock (see specification, page 3, lines 30-32; page 4, lines 26-31). The catalyst comprises a zeolitic material (see specification, page 6, lines 1-4) incorporating Al and one or more additional metals selected from the group consisting of Fe, Ga, Ti, and Co, and mixtures thereof (see specification, page 6, lines 5-10, page 9, lines 15-17), wherein the ratio of additional metal(s) is between about 1:10 and 2.5:1 (see specification, page 6, lines 6-10, page), and between 5 and 95 weight percent of a binder (see specification, page 10, lines 2-5).

Another invention is a process for preparing an isomorphically-substituted zeolitic catalyst (see specification, page 4, lines 5-7). The catalyst is prepared by selecting an aluminosilicate zeolitic material selected from the group consisting of ZSM-5, ZSM-11, ZSM-12, ZSM-22, MCM-22, ZSM-23, ZSM-39, ZSM-57, mordenite, Beta, FAU, and L-types (see specification, page 4, lines 5-9), and refluxing a slurry of the zeolitic material in the presence of a soluble metal

compound selected from the group consisting of metal compounds of Fe, Ga, Ti and Co and mixtures thereof under conditions effective for substituting aluminum in the aluminosilicate zeolitic material with metal from the soluble metal compound to yield a metal to aluminum ratio of between 1:10 and 3:1 (see specification, page 4, lines 9-13).

### Issues

The issues presented on appeal are:

- 1) Whether Claims 1-21 and 39-40 have been properly rejected under 35 U.S.C. 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter regarded as the invention?
- 2) Whether Claims 1-8, 10, 21-22, and 42 have been properly rejected under 35 U.S.C. 102 (b) as being anticipated by Iwatomo et. al (U.S. 5,207,893)?
- 3) Whether Claims 1,3, 5-6, 10-11, 14, 16-18, and 21 have been properly rejected under 35 U.S.C. 102 (b) as being anticipated by Suzuki et. Al (U.S. 4,994,254)?
- 4) Whether Claims 1-8, 10-11, 13-14, 16-19, 21-23, 25-26, 39, and 43 have been properly rejected under 35 U.S.C. 103(a) as being unpatentable over Skeels et. al (U.S. 5,098,687) in view of Farnos et. al. (U.S. 5,614,079) and further in view of Absil et. al (U.S. 4,837,397)?

### Grouping of Claims

Claims 1-8, 10, 11, 13-19, 21-23, 25, 26, 39, 40, 42, and 43 of the subject patent application stand or fall together.

### Argument

**I. Rejection of Claims 1-21 and 39-40 under 35 U.S.C. 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter regarded as the invention.**

The Appellants have addressed the 35 U.S.C. 112, second paragraph, rejection regarding claim 1 in the amendment submitted with this Appeal Brief. As for the rejections pertaining to claims 39 and 40, the Examiner has not

provided any reasons supporting the rejection. The Appellants do not see how they can provide a response to this rejection without any explanation given for the rejection of these claims.

**II. Rejection of Claims 1-8, 10, 21-22, and 42 under 35 U.S.C. 102 (b) as being anticipated by Iwatomo et. al (U.S. 5,207,893).**

In order for the claims of a prior art reference to anticipate a claim, the reference must teach every element of the claim. Iwamoto recites that its invention has 90-10% by weight of an inorganic oxide, and that such inorganic oxides include water containing oxides, alumina, and silica-alumina (see column 6, lines 4-7). However, claim 1 includes a recitation of between 5 to 95% weight percent of a binder. Iwamoto does not teach the inclusion of a binder in such a recited percentage range. Thus, Iwamoto does not teach each and every element of Claim 1 and its dependent claims. Iwamoto also recites for its process that "it is required that an iron salt is added to steam treated aluminosilicate to which mineral acid has been added, and in other words, the iron salt is added in the presence of the mineral acid" (see column 4, lines 65-70). However, the Appellants' invention recites "conditions effective for substituting aluminum in the aluminosilicate zeolitic material with metal from the soluble metal compound" in Claim 22. The Appellants' recite that metal compounds useful for isomorphic substitution reactions preferably are soluble metal fluorides such as iron trifluoride (see specification, page 7, lines 6-9). Iwamoto does not teach such conditions useful for isomorphic substitution of a portion of aluminum from a zeolitic starting material by another metal. As for the Examiner's statement that the "initial silica to alumina ratio increases following the treatment with the mineral acid and iron salt, which means that iron is substituted for alumina which has been removed from the framework," the Appellants' note the following inconsistency with that statement. The Examples 1 and 2 have a 3.5 weight percent of aqueous nitric acid, and have a silica to alumina ratio of 22.3 and 25.3, respectively. However, Example 3 has a 3.0 weight percent of aqueous nitric acid, but the silica to alumina ratio drops to 18.2, which is contrary to the Examiner's statement. In view of the preceding, the Appellants' respectfully request the honorable Board to reverse the Examiner's rejection of Claims 1-8, 10, 21-22, and 42 under 35 U.S.C. §102(b).

**III. Rejection of claims 1,3,5-6,10-11,14,16-18, and 21 under 35 U.S.C. 102(b) as being anticipated by Suzuki et al (U.S. 4,994,254).**

In order for the claims of a prior art reference to anticipate a claim, the reference must teach every element of the claim. The Examiner states that aluminogallosilicates having a ratio of 2-4 correspond to a Ga/Al ratio of 0.25-0.5. The Appellants' are not sure how the Examiner arrived at such a corresponding ratio computation. The Appellants' note Table 4 and the aluminogallosilicates recited therein. A computation of the Ga/Al ratio of the 17 samples provides a Ga/Al range of 0.036 to 2.78. By contrast, the Appellants' recite a ratio of between 1:10 at its lower end, and 2.5:1 at the higher end of the metal to Al ratio in its amended Claim 1. Suzuki does not teach the inclusion of a binder in such a recited percentage range. The Examiner also states that Suzuki recites aluminogallosilicates that may be formed into various shapes using alumina or silica binders, but there is no mention of any percent ranges of the binder as recited in the Appellants' invention. Also, the Examiner recites that Suzuki's composition may include an active metal such as palladium or platinum in an amount in the range of 0.1 to 10% by weight. By contrast, the Appellants' claims between 0.05 and 2 weight percent of a noble metal, and Suzuki does not teach a noble metal in such a recited percentage range. Regarding the process recitation of Claim 5, the Appellants' have provided reasons above to differentiate the aluminosilicate composition. The Appellants' respectfully requests the honorable Board to reverse the Examiner's rejection of Claims 1,3, 5-6, 10-11, 14, 16-18, and 21 under 35 U.S.C. §102(b).

**IV. Rejection of claims 1-8, 10-11, 13-14, 16-19, 21-23, 25-26, 39, and 43 under 35 U.S.C. 103(a) as being unpatentable over Skeels et. al (U.S. 5,098,687) in view of Farnos et. al. (U.S. 5,614,079) or Absil et al (U.S. 4,837,397).**

To establish a prima facie case of obviousness, three basic criteria must be met. First, there must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to modify the reference or to combine reference teachings. Second, there must be a reasonable expectation of success. Finally, the prior art reference (or references when combined) must teach or suggest all the claim limitations. The teaching or suggestion to make the claimed combination and the reasonable expectation of success must both be found in the prior art, not in

applicant's disclosure. *In re Vaeck*, 947 F.2d 488, 20 USPQ 2d 1438 (Fed. Cir. 1991).

Skeels discloses zeolite compositions which are topologically related to prior known zeolites but which are characterized as containing framework atoms of iron and/or titanium. By contrast, the Appellants' invention recites a zeolite material incorporation Al and one or more metals selected from the group consisting of Fe, Ga, Ti, Co, and mixtures thereof. Skeels does not teach metals such as Ga, Co, or mixtures of Fe, Ga, Ti, and Co.

The catalysts taught by the Appellants' are characterized by the presence of one or more additional metals in addition to those typically found in an aluminosilicate zeolitic lattice structure. The catalysts of the Appellants' invention contain significant amounts of both aluminum and one or more metals, including Fe, Ga, and/or Co. Furthermore, Skeels teaches catalysts including Group VIII noble metals alone, or in conjunction with Group IV-B metals in amounts between about 3 and about 15 weight percent of the overall catalyst composition. It would not have been obvious to a person of ordinary skill in the art to substitute iron in place of titanium in the ZSM-5 composition taught by Skeels. Likewise, incorporating the binders of Farnos and Absil would still not overcome the differences stated between the Appellants' invention and Skeels. Thus, the Appellants' respectfully request the honorable Board to reverse the Examiner's rejection of Claims 1-8, 10-11, 13-14, 16-19, 21-23, 25-26, 39, and 43 under 35 U.S.C. §103(a).

#### Conclusion

For the reasons stated above, the Appellants' submit that all claims now presented are in a condition for allowance, and respectfully requests the honorable Board to reverse the Examiner's final rejection of these claims.

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Respectfully submitted,



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## **Appendix of Claims**

1. A catalyst for methylating a naphthalenic feedstock, said catalyst comprising: a zeolitic material incorporating Al and one or more additional metals selected from the group consisting of Fe, Ga, Ti, and Co, and mixtures thereof, wherein the ratio of additional metal(s) is between about 1:10 and 2.5:1, and between 5 and 95 weight percent of a binder.
2. The catalyst of Claim 1 wherein there is a single additional metal, and the additional metal is Fe.
3. The catalyst of Claim 1 wherein at least fifty mole percent of the additional metal is incorporated into the zeolitic lattice structure.
4. The catalyst of Claim 3 incorporating a single additional metal, and the additional metal is Fe.
5. The catalyst of Claim 1 wherein the catalyst is a metallosilicate prepared by isomorphic substitution of Al by one or more additional metals.
6. The catalyst of Claim 5 wherein the metallosilicate is prepared by isomorphic substitution of Al by a single additional metal, and wherein the ratio of the single additional metal to Al is between about 0.16:1 and 2:9:1.
7. The catalyst of Claim 6 wherein the single additional metal is Fe.
8. The catalyst of Claim 7 wherein the ratio of Fe to Al is between 0.16:1 and 1.75:1.
10. The catalyst of Claim 1 wherein the binder is selected from the group consisting of binders comprising boehmite, alkali earth metals and SiO<sub>2</sub>.
11. The catalyst of Claim 1 further including between 0.01 and 5 weight percent of a noble metal.
13. The catalyst of Claim 11 wherein the noble metal is selected from the group consisting of platinum, palladium, rhodium, iridium and ruthenium, and wherein the catalyst includes a single additional metal, the single additional metal is Fe, and the ratio of Fe to Al is between 0.16:1 and 1.75:1.
14. The catalyst of Claim 1 wherein the weight percent of the noble metal is between 0.05 and 2.0 weight percent of the total catalyst weight.

15. The catalyst of Claim 7 wherein the catalyst includes 20 to 80 weight percent boehmite binder, wherein 0.5 to 2.0 weight percent of a noble metal is deposited on the binder, calculated as weight percent of binder plus noble metal.

16. The catalyst of Claim 1 where the zeolitic material is a ZSM-5 type aluminosilicate.

17. The catalyst of Claim 3 where the zeolitic material is a ZSM-5 type aluminosilicate.

18. The catalyst of Claim 5 where the zeolitic material is a ZSM-5 type aluminosilicate.

19. The catalyst of Claim 7 where the zeolitic material is a ZSM-5 type aluminosilicate.

21. The catalyst of Claim 1 wherein the zeolitic material is selected from the group consisting of ZSM-5, ZSM-11, ZSM-12, ZSM-22, MCM-22, ZSM-23, ZSM-39, ZSM-57, mordenite, Beta, FAU, L-types, and mixtures thereof.

22. A process for preparing an isomorphically substituted zeolitic catalyst comprising the steps of:

selecting an aluminosilicate zeolitic material selected from the group consisting of ZSM-5, ZSM-11, ZSM-12, ZSM-22, MCM-22, ZSM-23, ZSM-39, ZSM-57, mordenite, Beta, FAU, and L-types; and

refluxing a slurry of the zeolitic material in the presence of a soluble metal compound selected from the group consisting of metal compounds of Fe, Ga, Ti and Co and mixtures thereof under conditions effective for substituting aluminum in the aluminosilicate zeolitic material with metal from the soluble metal compound to yield a metal to aluminum ratio of between 1:10 and 3:1.

23. The process of Claim 22 wherein the refluxing is conducted in the presence of a soluble hydrogen fluoride salt.

25. The process of Claim 23 wherein the soluble metal compound is selected from the group consisting of metal fluoride compounds.

26. The process of Claim 22 wherein the zeolitic material is a ZSM-5 type material.



39. The catalyst of Claim 1 wherein a portion of acid catalyst sites in the catalyst have been intentionally deactivated prior to use of the catalyst in an alkylation reaction.

40. The catalyst of Claim 15 wherein a portion of acid catalyst sites in the catalyst have been intentionally deactivated prior to use of the catalyst in an alkylation reaction.

42. A catalyst formed by substituting Fe for Al in an alumino-silicate zeolytic matrix in which the Fe to Al ratio in a resulting metallosilicate matrix is from between about 1:10 to 3:1.

43. The catalyst of Claim 42 in which the alumino-silicate zeolytic matrix is of the ZSM-5 type.